

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Masanobu FUKUDA, et al.

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Examiner: Alexander C. Kollias

For: INK AND LAMINATED SHEET

DECLARATION UNDER 37 CFR §1.132

I, Masanobu FUKUDA, hereby declare and state that:

1. I am a citizen of Japan, residing at Komaki City, Aichi Prefecture, Japan.
2. I am one of the inventors of the subject application, and I am fully familiar with the subject matter thereof as well as the references relied upon by the Examiner in the prosecution of this application.
3. I entered Osaka University, Faculty of Engineering, Department of Engineering research, in April 1974, and graduated in March 1978. Furthermore, I entered the Graduate School of the Osaka University in April 1978, and graduated in March 1980 with a Master's Degree in Engineering.
4. I began working for Dainippon Ink and Chemicals, Inc., in April 1980. I was assigned to the resin technique fifth group of the resin technique headquarters of a Chiba Plant; assigned to the urethane technique group of the resin technique headquarters of a Sakai Plant in March, 1987; assigned to the crystalline liquid material technique headquarters of a Saitama Plant in October, 1998; assigned to the crystalline liquid material technique headquarters of Research Institute in April, 2000; assigned to

R & D headquarters of PX-1 project of a Warabi Plant in April, 2001; assigned to the new decorative technical first group of C & C headquarters of a Tokyo Plant in September, 2001; assigned to the decorative technical group of C & C headquarters of Research Institute in January, 2007; and assigned to the decorative technical group of Komaki Plant in May, 2009, and I have been engaged in developments of inks until now in Komaki Plant of DIC corporation. (Company name was changed from Dainippon Ink and Chemicals, Inc., to DIC corporation on 2008).

5. I conducted the following tests in order to show that the present invention can show excellent effects.

(Experiment)

Following experiments show that the present invention can show superior effect when the binder resin contains 50 to 500 mmol/kg of at least one selected from the group consisting of a carboxyl group, a phosphoric acid group, a sulfonic acid group and metal salts thereof. Since the effects of the range between 50 to 250 mmol/kg have already been shown in Examples of the present specification, the tests for showing the effects of the range between 250 to 500 mmol/kg were performed to show the effects of the present invention.

Preparation Example

(Polyester polyol)

2410.2 g of adipic acid (a product of Asahi Kasei Chemicals Corporation), 1589.8 g of 1,4-butanediol, 0.06 g of triphenylphosphine (Dislamoll TP manufactured by Bayer Corporation) were provided in a reaction vessel having a charging opening, which was equipped with a stirrer, a nitrogen introduce tube and a reflux condenser, and were gradually heated while suitable amount of nitrogen was supplied to the vessel. When the materials were melted and stirring became available, stirring was started. Dehydration condensation reaction thereof was performed for two hours at 140°C, two

hours at 220°C, and furthermore twelve hours at 220°C after a cooler condenser was removed, in this order, to obtain a white solid of polyester polyol A2 having an acid value of 0.4, hydroxyl value of 112, and the number average molecular weight of 1000.

(Preparation example 1 of polyurethane resin)

2002 g of the aforementioned polyester polyol A2, 134 g of 2,2-dimethylol propionic acid (abbreviated name: DMPA, a product manufactured by Nippon Kasei Chemical Co., Ltd), 0.8 g of dioctyltin dilaurate (a product manufactured by Nitto Kasei Co., Ltd.) and 1600 g of a mixture solvent (methyl ethyl ketone / cyclohexanone =1/1) were added in a reaction vessel having a charging opening, which was equipped with a stirrer, a nitrogen introduce tube and a reflux condenser. The mixture was heated at 80°C while stirring. After it was confirmed that the materials were melted and mixed uniformly, 265 g of hexamethylene diisocyanate (abbreviated name: HDI, a product manufactured by Mitsui Chemicals, Inc.) and 296 g of xylylene diisocyanate (abbreviated name: XDI, a product manufactured by Mitsui Chemical polyurethane, Inc.) were added to the mixture and reacted at 80°C. Solution viscosity of the mixture was measured, and when the solution viscosity of the resin solution arrived at the target viscosity, 10 g of methanol and 1333 g of methyl ethyl ketone were added to obtain a solution wherein urethane resin concentration of the solution was 45% by weight. The content of a functional group (carboxyl group) in the solid resin content was 310 mmol/kg (calculated value).

Similar to the preparation example 1 except that conditions were changed as shown in the following table, the preparation examples 2 was performed. Furthermore, preparation example 3 was performed in order to obtain a comparative polyurethane resin, which has the concentration of a functional group which exceed 500 mmol/g, such a resin was not obtained since gelation was occurred in the middle of the reaction.

| | Preparation Example 1 | Preparation Example 2 |
|--|--------------------------|--------------------------|
| Polyurethane resin | I | II |
| Polyester polyol A2 | 2002 g | 2039 g |
| DMPA | 134 g | 109 g |
| HDI | 265 g | 252 g |
| XDI | 296 g | 282 g |
| Content of a functional group (mmol/kg) | 417 g | 306 g |
| Non-volatile component (%) | 45.3 | 44.7 |
| Viscosity (mPas) | 9800 | 11500 |

(Preparation of ink)

Using the generated polyurethane resin I, an ink 11-A was prepared by mixing the following components. The concentration of aluminium foil pigment in the nonvolatile components of the ink 11-A was 35% by weight.

(Composition of an ink 11-A)

| | |
|--|----------|
| Aluminium foil pigment: "Starbrite (R) 4102EAC" manufactured by Siberline Corporation, ethyl acetate dispersion, solid content: 10%, | 30 parts |
| Binder resin: Polyurethane resin I (Non-volatile component 45%) | 12 parts |
| Ethyl acetate | 28 parts |
| Methyl ethyl ketone | 20 parts |
| Isopropanol | 10 parts |
| Epilcon B-4400 (manufactured by DIC corporation.) | 1 parts |

In addition, an ink 12-A was manufactured similar to the ink 11-A except that the aforementioned polyurethane resin II was used instead of the polyurethane resin I.

Example 1 and 2

Using the ink 11-A and the ink 12-A, the layer constitution was prepared as follows.

The layer constitution of film A/ink 11-A or 12-A/adhesive D-1/film C was prepared such that the ink 11-A or ink 12-A was applied in a dry thickness of 2.0 μm using a reverse gravure coater and the adhesive D-1 was applied in a coating weight of 5.0 g/m^2 using a gravure coater.

Film A: Technolloy R 526, manufactured by Sumitomo Chemical Co., Ltd., the thickness thereof is 125 μm

Adhesive D-1: the adhesive D-1 was prepared according to the method for an adhesive D-1 described in the present specification.

Film C: a gray and opaque ABS film having a thickness of 300 μm

The resulting sheet was aged at 40°C for 3 days, and then the surface luster value and the peel strength were measured. The surface luster value was measured under the conditions of 60°/60° using a glossmeter; MICRO-TRI-GLOSS (manufactured by BYK-Gardner). The peel strength (N/in) was measured by T-peeling of a film A and a film C.

Furthermore, using the inks 11-A and 12-A, each residual ink after used for gravure coating was stored in a sealed constant temperature bath at 40°C for one week and the state of the ink was examined. The viscosity was determined by measuring the time in seconds required for dropping an ink at room temperature of 23°C using a Zahn cup No. 3 manufactured by RIGO Co., Ltd. The results are shown in the following Table 1.

Table 1

| Ink | Surface luster value | Interlaminar peel strength (N/in) | Viscosity upon preparation (sec) | Viscosity after aging at 40°C for 7 days |
|----------|----------------------|-----------------------------------|----------------------------------|--|
| Ink 11-A | 170 | 7.7 | 21 | 109 |
| Ink 12-A | 157 | 7.3 | 22 | 79 |

These results show that the present invention can provide a high-brightness ink which imparts high-brightness mirror-like metallic luster for the printed surface and also has high peel strength.

6. I understand fully the content of this declaration.

7. I, Masanobu FUKUDA, the undersigned declarant, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Masanobu Fukuda

Masanobu FUKUDA

Date: Nov. 11. 2010